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Some Computations of Long-Distance
Drift of Aerial Sprays

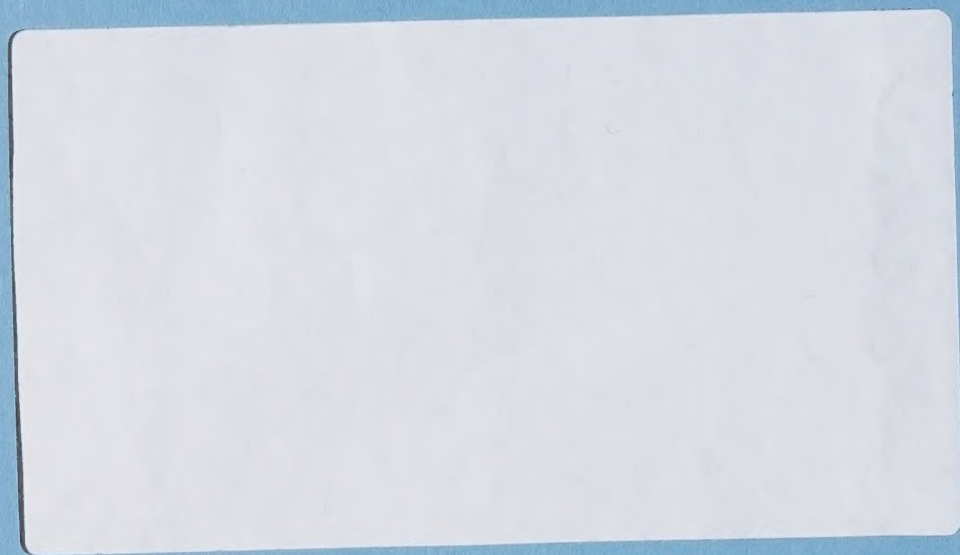
by

John D. Reid

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Some Computations of Long-Distance
Drift of Aerial Sprays

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John D. Reid

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Abstract

Spraying of 25 minutes duration by 3 TBM tankers of 600 US gallon capacity each over 3.2 km (2 miles) long tracks normal to the wind is investigated using a Monte Carlo trajectory method. Effective spray release height is 6 m above tree top. A weak wind, 2 m s^{-1} at 10 m above the spruce forest canopy, is assumed with a neutrally stratified lower atmosphere capped by an inversion at 400 m.

For the spray formulation considered (11% Fenitrothion, 1.5% Dowanol Solvent, 1.5% Atlox Emulsifer, 86% Water) evaporation of the aqueous component proceeds rapidly at ambient 65% relative humidity. Within 15 seconds of generation evaporation is effectively complete and the mass weighted droplet spectrum mode diameter has shifted to $\sim 52 \text{ }\mu\text{m}$, half its original value.

Within the first 500 m downwind of spraying, about 82% of the Fenitrothion residual moves below tree-top height and is assumed deposited. Since larger drops have greater settling velocities deposits are dominated by the larger droplets and the spectrum of airborne droplets moves to smaller sizes. By 80 km (50 miles) downwind the mass weighted spectrum mode diameter is at $21 \text{ }\mu\text{m}$, a very small mass ($\sim 0.006 \text{ g}$) is in vapour and droplets to $2 \text{ }\mu\text{m}$ diameter, a little over 1 kg is in droplets from 2-25 μm diameter, and about 1/3 kg in droplets larger than $25 \text{ }\mu\text{m}$.

The highest surface concentration at 80 km downwind is approximately $0.05 \text{ }\mu\text{g m}^{-3}$, averaged horizontally over 500 m along the wind and vertically over 38.8 m from the surface up, and occurs about 5 3/4 hours after spraying starts. Concentrations of at least one-tenth

the maximum are found between 25 minutes before and 20 minutes after the passage of the maximum.

Computation results are strongly influenced by the form of the initial droplet spectrum, droplet evaporative behaviour and effective source height. The neglect of re-emergence of droplets and vapour from the forest canopy could also be important. Of the meteorological factors, wind direction is critical where a particular location is of concern; atmospheric stability and wind speed are also important. Finally, the effects of chemical transformations should not be overlooked.

1. Introduction

Pesticide spraying to reduce the impacts of the Spruce Budworm moth has now been conducted in New Brunswick forests for 25 years. Such spraying is designed to kill the larvae in the immediate spray area. Recently concern has grown that an important fraction of the toxic spray escapes the target forest area and drifts considerable distances to very different environments.

At the request of the Government of New Brunswick, through the University of New Brunswick, the Atmospheric Environment Service has conducted an evaluation of the potential for such long-range transport using a predictive modelling approach. In order to focus the discussion of this problem by a number of different groups a spray scenario was formulated. With different groups employing different assessment technologies the objectives were: to identify the extent of the problem; recommend on the desirability of, and form of, a monitoring program and; identify areas of poor understanding which cause important errors in the various assessments.

This report documents the assessment prepared by the Atmospheric Environment Service. The technique employed is that of tracing the trajectories of typical spray droplets under the influence of the different processes acting. In the following section the spray scenario is detailed. Then the basis for the representation of each process, and potential errors are presented in section 3. This is followed by a brief consideration of the model computational procedure and detailed discussion of model results and conclusions.

2. The Spray Scenario

Geographically the area of concern is in Northern New Brunswick in a region cut by the Nepisiquit River Valley. However, for the purposes of this study the influences of topography are neglected. Pesticide spraying is assumed to occur in an area 50 miles (80 Km) directly upwind of the downwind target of interest, the Allardville Fire Tower. A second target at Bathurst, about 25 Km north of Allardville, is also of interest. The area is assumed covered by forest with a tree height of 40 feet (12 metres).

Aerial spraying is assumed to take place by three TBM tankers, each with 600 U.S. gallons capacity. They are assumed to spray in formation back and forth along a 2 mile (3.2 Km) line normal to the wind. Spraying at 1 U.S. gallon per minute through each of 24 nozzles. The total spray endurance is assumed to be 25 minutes, starting at 6 a.m.

Composition of the spray is, by volume:

- 11% Fenitrothion
- 1.5% Dowanol Solvent
- 1.5% Atlox Emulsifer
- 86% Water

The Fenitrothion and Atlox are found not to evaporate perceptably so the vapour pressure for spray evaporation is taken to be that for pure water down to 12.5% of the original volume.

The droplets initially produced are assumed to have a Gamma distribution with mass mean diameter $82\mu\text{m}$. The cumulative number and mass distributions are shown in Figure 1. Density of the Fenitrothion residual is 1.33 g cm^{-3} . The effective spray emission height above tree top level is 20 feet (6 metres). This accounts for the effects

of aircraft trailing vortices on the initial spray motion and any bulk sedimentation effects.

Meteorologically there is assumed to be a neutral stability layer from the surface to 400 metres above ground, where there the layer is capped by an inversion. Winds are assumed from the west, with friction velocity 0.3 m s^{-1} , roughness length 1 metre and displacement height 90% of tree height or 35 feet (11 metres). This gives a wind speed of 2 m s^{-1} at 10 m above tree-top and 5 m s^{-1} near inversion base.

Finally, and to avoid the problems of dealing with in-canopy transport processes, any droplets which move beneath tree-top level are treated as trapped and not considered further.

3. Physical Processes

Numerous interacting physical processes pertain to the fate of aerial sprays. Their relative importance depends upon the specific spray in question (e.g. chemical composition, spectrum of drop sizes), and the conditions of spraying (e.g. spray height, meteorological conditions). Specific examples are: presence of cloud during the day-time can be important for photo-reactive spray; occurrence of rain can lead to rapid sweeping out of large quantities of spray; sea breezes and other local flow phenomena can lead to long distance drift only weakly related to wind at the spray source location. A good general treatment allowing for all possible processes and interactions is far beyond the bounds of current knowledge. Moreover, a crippling limiting factor would be the large amount of computation needed.

In the next few sections the specific processes treated in this study, transport by the mean wind, vertical turbulent motion, crosswind dispersion, sedimentation, evaporation and surface deposition are considered in detail.

3.1 Transport by the Mean Wind

Wind varies on a variety of temporal scales. For convenience of treatment in meteorology two ranges of scales are defined. The mean wind covers the larger scales and may vary with respect to position, time and height but is considered to be explicitly definable in each dimension. Smaller scales of fluctuation, turbulence, can only be defined in terms of statistics of fluctuations. The effects of these turbulent scales of motion are treated in the following two sections.

The mean wind components are the results of the flow produced by synoptic migratory weather systems and flows produced (or modified) by features of the local physical geography (topographic channelling, sea breezes, etc.). In general, for the accuracies needed in case studies the prediction of the mean flow by numerical integration of the equations of motion is not satisfactory and actual observations are employed to diagnose the mean flow field. For short range trajectories the mean wind is often assumed to have no horizontal variation and the effects of vertical variation are incorporated into the treatment of turbulent transport effects. For longer range transport an analysis of distributed observations can be conducted; a good example is the MATHEW model of Sherman (1978).

In many cases wind observations are only available at the surface. Assumptions are then necessary about the vertical variation. Over homogeneous surfaces, such as grass prairie, observations show logarithmic or log-linear wind speed increase with height in the lowest few tens of metres depending on atmospheric stability, with constant wind direction. According

to Mukammal (personal communication) profiles measured over forests do not tend to show such regularity, probably owing to surface inhomogenities. Forest profiles do not appear to have been systematized. This being the case it is reasonable to maintain a rather simple approach and assume a logarithmic wind profile

$$u(z) = \frac{u_*}{k} \ln \left(\frac{z-d}{z_0} \right)$$

where z_0 is roughness length (1 m), d is displacement height (11 m), u_* is friction velocity (0.3 m s^{-1}) and k is von Karman's constant (0.35).

Above the lowest few tens of metres wind speeds usually continue to increase and direction often varies consistently from that at the surface. Observational data become indispensable where trajectories pass into this region. For this scenario calculation there is probably little better alternative than to assume a continuation of the surface layer profile.

3.2 Vertical Turbulent Motion

Particles move vertically owing to the action of vertical turbulent air motion. The trajectory approach to non-buoyant particle dispersion has been investigated by Reid (1978, 1979) using Monte Carlo techniques. Particle displacements are computed for time intervals Δt using

$$\Delta z = w' \Delta t$$

where w' is the instantaneous particle vertical velocity. w' values at successive times are assumed given by a Markov Chain relation

$$w'(t+\Delta t) = \alpha w'(t) + \beta r$$

Here r is a random number drawn from a normal distribution $N(0, \sigma_w)$ σ_w is the standard deviation of the turbulent wind vertical velocity distribution at the level of the particle, and α and β are derived from the turbulence statistics. From the requirement for constancy of $\overline{w'^2}$ (the particle turbulent vertical velocity variance) in a uniform σ_w field

$$\beta = (1 - \alpha^2)^{1/2}$$

By assuming that the autocorrelation

$$\frac{\overline{w'(t)w'(t+\Delta t)}}{\overline{w'^2}} = \exp\left(-\frac{\Delta t}{T_L}\right)$$

an exponential decay with memory T_L (the Lagrangian time scale),

$$\alpha = \exp\left(-\frac{\Delta t}{T_L}\right).$$

A recent paper by Neumann (1978) confirms the utility of the exponential assumption.

In this study σ_w^2 is assumed given by its surface layer value

$$\sigma_w^2 = 1.56 u_*^2$$

throughout the sub-inversion layer depth. Deardorff (1972) suggests that σ_w decreases slowly with height in neutral conditions above the surface layer, but the vertical variation is not well established and the assumption of invariance is not unreasonable.

Even less certainty exists about the variation of Lagrangian time scale. Reid (1979) argues for a relationship

$$T_L = 0.4 \frac{z}{u_*}$$

for the neutral surface layer which gives good agreement between dispersion simulations and observations. Neumann suggests $T_L \sim 70$ sec for the neutral boundary layer. In this study Reid's expression is modified to

$$T_L = 0.4 \frac{(z-d)}{u_*} \exp \left(-3 \frac{(z-d)}{h} \right)$$

where h is inversion height and d is incorporated to account for displacement height effects. Thus T_L has a maximum at $\sim 1/3$ inversion height giving an asymptotic vertical eddy diffusivity profile, $K_w = \sigma_w^2 T_L$, with conventional behaviour. For the parameters of this scenario, $u_* = 0.3 \text{ m s}^{-1}$, $h = 400 \text{ m}$, the maximum T_L is ~ 65 seconds and average $T_L \sim 45$ seconds.

In calculating trajectories for particles with non-zero settling velocities the effect of particles falling through the turbulence field, rather than being completely advected along with it need to be accounted for. Csanady (1963) considered this trajectory crossing effect and showed that it is not important for small settling velocities. Since only small particles (droplets) having small terminal velocities are able to remain in the atmosphere to the downwind distances of interest here, this effect is not incorporated in these calculations.

3.3 Crosswind Dispersion

The effects of lateral turbulent velocity fluctuations, and to a limited extent the climatological vertical variation of wind direction, are taken into account using results of the Gaussian plume model. This model assumes that the time mean concentration (χ) profiles (vertical and lateral) from a continuous point source at $x = y = z = 0$

can be described by an expression of the form

$$\chi(x, y, z) = \frac{Q}{2\pi\bar{u}\sigma_y\sigma_z} \exp\left(-\frac{1}{2}\left(\frac{y^2}{\sigma_y^2} + \frac{z^2}{\sigma_z^2}\right)\right)$$

Here Q is pollution source strength, $\bar{u}$ is mean wind velocity, and $\sigma_y(x)$, $\sigma_z(x)$ are the crosswind and vertical pollutant distribution standard deviations respectively. The expression is much used in air quality assessments with numerous modifications being introduced to account for non-standard conditions.

In the spray scenario the special modification needed in treating the effect of lateral dispersion is to account for the source being a line source of finite length, not a point source. According to the scenario spray tracks are 2 miles (3.2 km) long and flown normal to the wind. Assuming a surface level concentration and source ($z=0$), the Gaussian equation can be integrated to give the concentration at (x, y) , where y is measured from the centre at the spray track

$$\chi(x, y) = \frac{Q}{\sqrt{2\pi}\bar{u}\sigma_z} \left[\operatorname{erf}\left(\frac{y+s}{\sqrt{2}\sigma_y}\right) - \operatorname{erf}\left(\frac{y-s}{\sqrt{2}\sigma_y}\right) \right]$$

Here s is the half length of the spray track (1 mile).

In the scenario the downwind distance of interest is ~ 80 Km and for this distance and neutral stability $\sigma_y \approx 3.4$ Km. For ease of computation assume $\sigma_y = 2$ s. For these conditions Figure 2 shows the crosswind variation, normalized as y/s , of concentration, plotted as a fraction of concentration at the same distance downwind of an infinite cross-wind line source. Notice that the axial concentration fraction

is only 0.38 and decreases rapidly. At $y = 16$ s the concentration fraction is down to $\sim 5 \times 10^{-15}$. Clearly the off axis concentrations (at Bathurst in the scenario) are of negligible concern relative to the axial ones. Nevertheless, it is important to appreciate that any specific wind direction will put a given geographic location on the axis.

3.4 Sedimentation

Gravitational settling of spray droplets is treated as fall at the terminal velocity appropriate to a sphere of the same density. Approximate calculations show that even the slowest responding droplets (the largest) for the scenario reach terminal velocity from rest in a fraction of a second. The effects of non-sphericity also would seem to be insignificant judging by the data of Berry and Pranger (1974). An efficient method for computing terminal velocity which accounts for deviations from Stokes law drag is to calculate the Best number (X), the product of the drag coefficient and Reynolds number squared, which is related to droplet mass, air density and dynamic viscosity by

$$X = \frac{8}{\pi} \frac{mg\rho_a}{\eta_d^2}$$

the Best number is then a monotonic function of Reynolds number, over the range of concern, and empirically given by a polynomial

$$Re = a_{12}X + a_{12}X^2 + a_{13}X^3 + a_{14}X^4$$

Following Berry and Pranger (1974) the values

$$\begin{aligned} a_{11} &= 0.412657 \times 10^{-1} \\ a_{12} &= -0.150074 \times 10^{-3} \\ a_{13} &= 0.758804 \times 10^{-6} \\ a_{14} &= -0.168841 \times 10^{-8} \end{aligned}$$

good for the range of Reynolds number encountered, are applied. The terminal velocity is then computed from

$$V_T = \frac{\eta_d R_e}{2r\rho_a}$$

Figure 3 shows terminal velocity as a function of droplet diameter.

In the simulation, with only the water component of the droplets evaporating, the bulk density of the droplet increases as it evaporates. The effect on terminal velocity is taken into account and shown in Figure 3.

3.5 Evaporation

Evaporation of the airborne spray droplets is treated as a mass transfer type process following the formulation of Goering et al (1972). They give the rate of diameter change of an evaporating spray droplet as:

$$\frac{dD}{dt} = -2 \left(\frac{M_v}{M_m} \right) \left(\frac{D_v}{D} \right) \left(\frac{\rho_a}{p_l} \right) \left(\frac{\Delta p}{p_f} \right) (2 + 0.6 Sc^{1/3} Re^{1/2})$$

Each of the terms is evaluated at an ambient temperature of 10°C so that:

M_v is molecular weight of the diffusing water vapour from the drop (18);

M_m is mean molecular weight of gas mixture in the transfer path (29);

D_v is the diffusivity of water vapour in air ($0.241 \text{ cm}^2 \text{ s}^{-1}$);

ρ_a is air density ($1.23 \times 10^{-3} \text{ g cm}^{-3}$);

p_l is density of the droplet. This varies as water evaporates and the Fenitrothion of greater density is a greater proportion of the residual ($1.04 - 1.33 \text{ g cm}^{-3}$);

p_f is atmospheric pressure (partial pressure of air) (760 mm Hg);

Δp is the difference between the saturation vapour pressure at the surface of the drop and the vapour pressure in the surrounding atmosphere. For droplet sizes in question the curvature of surface effect on surface vapour pressure is negligible. The effect of presence of dissolved substance is also neglected. At ambient relative humidity of 60% $\Delta p = 3.68 \text{ mm Hg}$;

Sc is the Schmidt Number

$$Sc = D_v / \eta_k$$

where η_k is kinematic viscosity of air ($0.144 \text{ cm}^2 \text{ s}^{-1}$);

Re is the Reynolds Number

$$Re = W_a D / \eta_k$$

where W_a is velocity of the droplet relative to the air (droplet terminal velocity as computed in section 3.4).

These conditions were entered into a preliminary calculation and it was determined that droplets evaporated very quickly down to half their original diameter (12½% original volume). For example, an original 128 μm diameter droplet evaporated to 64 μm in ~12 seconds. Smaller droplets evaporate even more rapidly. By the assumptions of this scenario no evaporation of Fenitrothion residual is allowed. It was decided that for this scenario evaporation could be considered to occur instantaneously after spraying without incurring any appreciable error. Evaporated water vapour is of no further interest. Figure 4 shows the effect on the mass distribution of spray droplets. The curve labelled "Original Spray" is just the derivative of the curve labelled "Mass" in Figure 1. The mode of this distribution is at ~104 μm with the ordinate normalized to unity at the mode. The evaporated spray droplet distribution has a mode of ~52 μm . However, notice that because

of the shape of the spectrum and greater density of evaporated droplets there is actually greater mass of droplets smaller than $\sim 45 \mu\text{m}$ than originally. In fact, for all droplets smaller than $2 \mu\text{m}$ diameter the mass has increased from $\sim 0.007 \text{ g}$ to $\sim 0.067 \text{ g}$, for droplets from 2 to $25 \mu\text{m}$ diameter the increase is from 10.53 to 40.79 kg , and for 25 to $50 \mu\text{m}$ diameter droplets the increase is from 242.2 to 354.5 kg . Only for droplets larger than $50 \mu\text{m}$ does the mass decrease, from 6765 to 737.5 kg .

3.6 Deposition

A full treatment of this spray situation would require accounting for flow, dispersion and deposition effects within the forest canopy. Understanding of these processes in the forest is poor and there treatment must be regarded as beyond the scope of this study. As a great simplification the problem is circumvented by specifying that any droplet or vapour molecule which moves below tree-top level is deposited.

Chamberlain (1975) gives a good review of vapour and droplet deposition. Generally large particles and moist particles are efficiently deposited but small particles and vapours are less efficiently deposited. Deposition is greater to moist and sticky surfaces. The perfect deposit efficiency assumed in this study will tend to overestimate deposition of vapour and small particles, but as these contain only a small proportion of the mass this will not likely constitute a serious error.

Another implication of this treatment of deposition is that the pesticide cloud will tend to pass a particular location more rapidly in the simulation than in reality. By neglecting in-canopy processes

delays which occur when particles enter slowly moving air within the canopy and then re-emerge are not taken into account. This effect will also, at least to a first order approximation, lead the model to predict higher concentrations than actually occur. The time delay can be looked upon as extending the range of scales of lateral velocity fluctuations of significance. This will tend to lead the model to over-estimate axial deposition.

One further process which is neglected, but is certainly likely to be significant, is evaporation of deposited insecticide. This will act as a distributed secondary vapour source of low intensity but prolonged duration. Insecticides with long chemical half-lives will be the most important.

4. Computations

4.1 Computational Procedures

Separate computations were performed for representative droplet sizes across the spectrum of sizes for evaporated spray at 0 Km (Figure 4). As this computational technique was under development the computations were performed in two stages: - (i) computations of fraction of particles surviving in the atmosphere at different downdwind distances; (ii) computations of insecticide cloud concentrations. The separation is useful for development and is maintained for clarity in this discussion, but the computations can in fact be performed together with considerable saving in computational time.

In each of these computations droplet trajectories are calculated for small time steps under the influence of vertical turbulent motion, gravitational settling and mean horizontal winds with the assumption

that each of these influences remains constant for the course of one time step. Time steps of 20% of the Lagrangian time scale are adopted except near the surface where a lower limit of 1 second is established. Numerous tests showed that results were not systematically adversely influenced by the choice of time step here.

In this type of computation it is always necessary to specify the fate of a droplet which encounters an upper or lower boundary. The assumption of complete deposition at tree-top level has already been explained in section 3.6. In the computation this is implemented as just going on to the next droplet trajectory computation when the ground is reached. For the upper boundary, which is an inversion at 400 m in this computation, a perfect reflection condition is assumed. If a particle is above the inversion at the end of a time step it is moved an equal distance below and the sign of its turbulent velocity is reversed.

Computations of fraction of droplets remaining in the atmosphere were made by noting the passage of droplets past each 1/2 km distance from the source. The number of droplets still airborne as a fraction of the total number released is the appropriate fraction for that particular size and distance. Up to 10,000 initial droplets were followed, although for those with small settling velocities this required large amounts of computer time. A workable solution to this time problem was to compute for about 500 octal seconds (daytime run limit on the AES CDC 7600 computer). This way, computations for the large droplets which settled out early included a large number of initial droplets, while for slowly settling droplets fewer (as few as 1500)

could be followed, but since these survived in the atmosphere to greater downwind distances, definition of the airborne pesticide residual was still satisfactory. These computations are useful for examining surface deposition since, by continuity, the difference in airborne fraction between two downwind distances can only be accounted for by deposition.

For computing concentrations the atmosphere is divided into cells which are the ultimate resolution elements for concentration computations. One thousand droplets are released from the source at the initial time and moved, time-step by time-step through the atmosphere for a 25 minute time segment. The sum of the times that droplets spend in each cell is proportional to concentration in that cell. The co-ordinates and turbulent velocities of each droplet are stored for use in computing the next time segment. A time segment length of 25 minutes is selected as it corresponds to the spray time from the aircraft. The cell concentrations found after each time segment correspond to the ensemble mean instantaneous concentrations one would expect to observe at 25 minute increments. As droplets settle out the number remaining in the atmosphere becomes small and the concentration pattern begins to look patchy. To overcome this problem a technique known as cloning is adopted. When, at the end of a time segment, the number of airborne droplets is less than half the number originally released a new twin droplet is created for each old one with all the properties of the old one. However, each of the twin droplets are assigned only half the weight in contributing to concentration that the one original droplet had. Computations proceed treating each of the twins separately and because their trajectories

are influenced by different random numbers the twins diverge. This cloning process can be repeated as further deposition occurs.

In each of these two computational procedures different droplet sizes are examined, for example, 0 μm , 5 μm , 15 μm , 25 μm , 40 μm and 60 μm diameter droplets were taken for one series of computations. The downwind mass fraction (or concentration) computed can then be plotted against diameter and interpolation used to derive values for intermediate diameters. This is done by weighting with the mass in the original mass distribution spectrum (after evaporation) and using numerical integration to compute the mass (or concentration) for the whole spectrum of droplet sizes.

4.2 Results and Conclusions

Figure 5 shows some results of these calculations in terms of percent of droplets, of various terminal velocities, remaining in the atmosphere for downwind distances to 100 km. Terminal velocities may be related to droplet diameter by referring to Figure 3. Two features of the evolution are of particular note:

- (i) The initial deposition is large. Detailed calculations show that $\sim 82\%$ of residual Fenitrothion mass is deposited within 500 m of spraying. A breakdown of percent of drops deposited, by terminal velocity (drop-size), is included in Table 1. Note that only about 50% of the smallest droplets are deposited in this target region. Subsidiary calculations show that the effective spray height has a strong influence on target area deposition of small droplets.

(ii) The fraction of droplets deposited declines markedly downwind, particularly for the larger droplets. For example, Table 1 shows at 9.5 to 10 km downwind the percent deposition at vapour is $\sim 10\%$ of its value for the first 500 m, whereas for -10 cm s^{-1} fall velocity it is only $\sim 0.1\%$. Altogether only $\sim 0.1\%$ of the Fenitrothion mass at spraying is deposited between 9.5 and 10 km.

Notice in Figure 5 that there is some statistical scatter in the calculations through which smooth curves have been drawn. Data for two calculations at -5 cm s^{-1} have been included to illustrate the scatter introduced by using different random number generator seeds. The scatter does not appear to be significant within the context of the representativeness of the scenario and validity of the parameterizations.

Figure 6 shows the spectrum of airborne droplets at 80 km, produced by applying the percentages at 80 km in Figure 5 to the ordinate values for the evaporated spectrum in Figure 4. Note the expanded ordinate scale in Figure 6. The airborne spectrum mode has shifted to $21 \mu\text{m}$ with larger droplets having had a preferential tendency for deposition. Table 2 shows the breakdown of mass in different regions of the spectrum. A little over 1 kg of insecticide residual is in the 2-25 μm class and about 1/3 kg in the 25-50 μm class. This represents about 0.12% of the original insecticide mass.

Concentration calculations were performed for 0, 5, 15, 25, 40 and 60 μm diameter droplets. The time of maximum concentration at 80 km was found to depend slightly on droplet diameter. The variation, between 5 hr 42 minutes and 5 hr 50 minutes after the start of spraying, is not significant. A similar slight variation occurs in the times concentrations

at 80 km first and last attain one-tenth the maximum value. The onset was about 25 minutes before and the end about 20 minutes after passage of the maximum

Applying the breakout for lateral dispersion of section 3.3, maximum concentration averaged over a cell at 80 km having dimensions 500 m along the wind and 38.8 m vertically from the surface up was computed to be $0.05 \mu\text{g m}^{-3}$. Mass contributions from droplets greater than and less than $25 \mu\text{m}$ were about equal, reflecting a tendency for larger droplets to be found in the lower part of the atmosphere. As noted previously concentrations decrease rapidly with off-axis distance.

Maximum concentration is particularly sensitive to the mass of material originating in the smaller drop sizes. For example, an additional 10 kg of pesticide in 0 to $5 \mu\text{m}$ diameter droplets would increase maximum surface concentration by $\sim 0.02 \mu\text{g m}^{-3}$; this would represent a shift of less than 1% of the residual Fenitrothion mass from larger droplet sizes.

In considering the implications of the results of this study it is important to appreciate that certain conditions of the scenario may not be necessarily very representative of actual spraying conditions. Best guesses were made for conditions and parameterizations where these are not yet well defined by measurements. As a consequence not too much attention should be paid to absolute values reported. The simulations are much more useful in defining trends and the relative importance of the interacting processes.

The computation results are strongly influenced by the form of the initial droplet spectrum and droplet evaporative behaviour. In

general, the greater the mass sprayed, or evaporated rapidly into the smallest droplet sizes (and vapour), the greater the long distance drift will be. Current lack of knowledge of the mass distribution within the lowest 30 μm droplet diameter range is a significantly limiting factor for fidelity in simulation. Likewise, effective emission height is important in governing the time available until larger droplets impact on the surface. Neglect of evaporation from deposited spray is another important limitation of this simulation. A regional survey by Yule et al (1971) showed elevated background phosphorus* levels in the New Brunswick atmosphere during the spray season which suggest the importance of this secondary source.

Of meteorological factors, wind direction is critical where a specific location is of concern. This is particularly so because of its wide diurnal and spatial variations. Stability variations, resulting in turbulence variations, also need to be well defined. Specifically, the assumption of low-level neutral stratification here is not likely very representative of a more typical variation from stable during spraying, through neutral, to unstable later in the morning.

Lastly there are some broader issues that need to be addressed. What are the appropriate interface variables between this type of atmospheric simulation and the biological receptors of concern? Is it instantaneous pesticide concentrations, longer term average concentrations or dosages which are of concern? Or is it perhaps that one stray spray droplet in the wrong place is the most environmentally significant?

* Phosphorus is a component of Fenitrothion

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Sample No.	Date	Time	Wind Dir.	Wind Spd.	Temp.	Humid.	Pressure	Fog	Clouds	Precip.	Fenitrothion (ppm)	
											Atmosphere	Soil
1	10/10/71	10:00	100	10	65	65	30.0	0	0	0	0.1	0.1
2	10/10/71	11:00	100	10	65	65	30.0	0	0	0	0.1	0.1
3	10/10/71	12:00	100	10	65	65	30.0	0	0	0	0.1	0.1
4	10/10/71	13:00	100	10	65	65	30.0	0	0	0	0.1	0.1
5	10/10/71	14:00	100	10	65	65	30.0	0	0	0	0.1	0.1
6	10/10/71	15:00	100	10	65	65	30.0	0	0	0	0.1	0.1
7	10/10/71	16:00	100	10	65	65	30.0	0	0	0	0.1	0.1
8	10/10/71	17:00	100	10	65	65	30.0	0	0	0	0.1	0.1
9	10/10/71	18:00	100	10	65	65	30.0	0	0	0	0.1	0.1
10	10/10/71	19:00	100	10	65	65	30.0	0	0	0	0.1	0.1
11	10/10/71	20:00	100	10	65	65	30.0	0	0	0	0.1	0.1
12	10/10/71	21:00	100	10	65	65	30.0	0	0	0	0.1	0.1
13	10/10/71	22:00	100	10	65	65	30.0	0	0	0	0.1	0.1
14	10/10/71	23:00	100	10	65	65	30.0	0	0	0	0.1	0.1
15	10/10/71	00:00	100	10	65	65	30.0	0	0	0	0.1	0.1
16	10/10/71	01:00	100	10	65	65	30.0	0	0	0	0.1	0.1
17	10/10/71	02:00	100	10	65	65	30.0	0	0	0	0.1	0.1
18	10/10/71	03:00	100	10	65	65	30.0	0	0	0	0.1	0.1
19	10/10/71	04:00	100	10	65	65	30.0	0	0	0	0.1	0.1
20	10/10/71	05:00	100	10	65	65	30.0	0	0	0	0.1	0.1

Table 1: Percentage of droplets, of various terminal velocities
(diameters), deposited from 0-0.5 and 9.5-10.0 km downwind.

Distance Range, km	Terminal Velocity cm s^{-1} (Droplet Diameter, μm)	0	-1.0	-2.5	-5.0	-10.0	-15.6	-30.0	-50.0
		(0)	(16)	(25)	(35)	(50)	(64)	(94)	(130)
0 - 0.5		43.8	48.9	53.8	62.0	77.6	88.9	98.9	99.98
9.5 - 10.0		0.47	0.56	0.49	0.39	0.10	0.04	0	0

Table 2: Airborne mass budget for various droplet size ranges

Diameter Range (μm)	Original Mass (kg)	Evaporated Mass (kg)	Mass at 80 km (kg)
0 - 2	7.02×10^{-6}	6.68×10^{-5}	6.01×10^{-6}
2 - 25	10.53	40.79	1.04
25 - 50	242.2	354.5	0.34
>50	6765.0	737.5	-

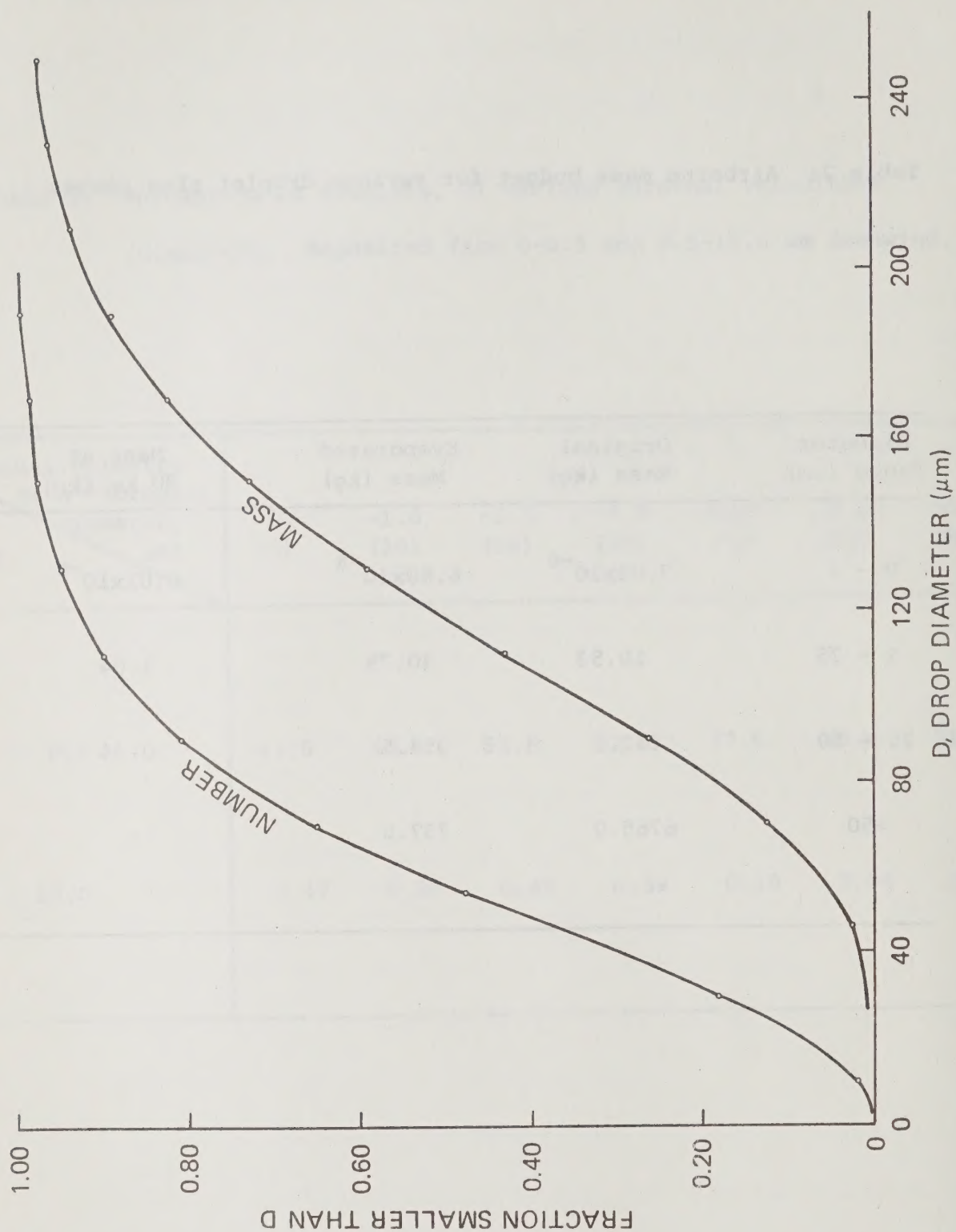


Figure 1: Cumulative Number and Mass Distributions for the Initial Spray Droplets

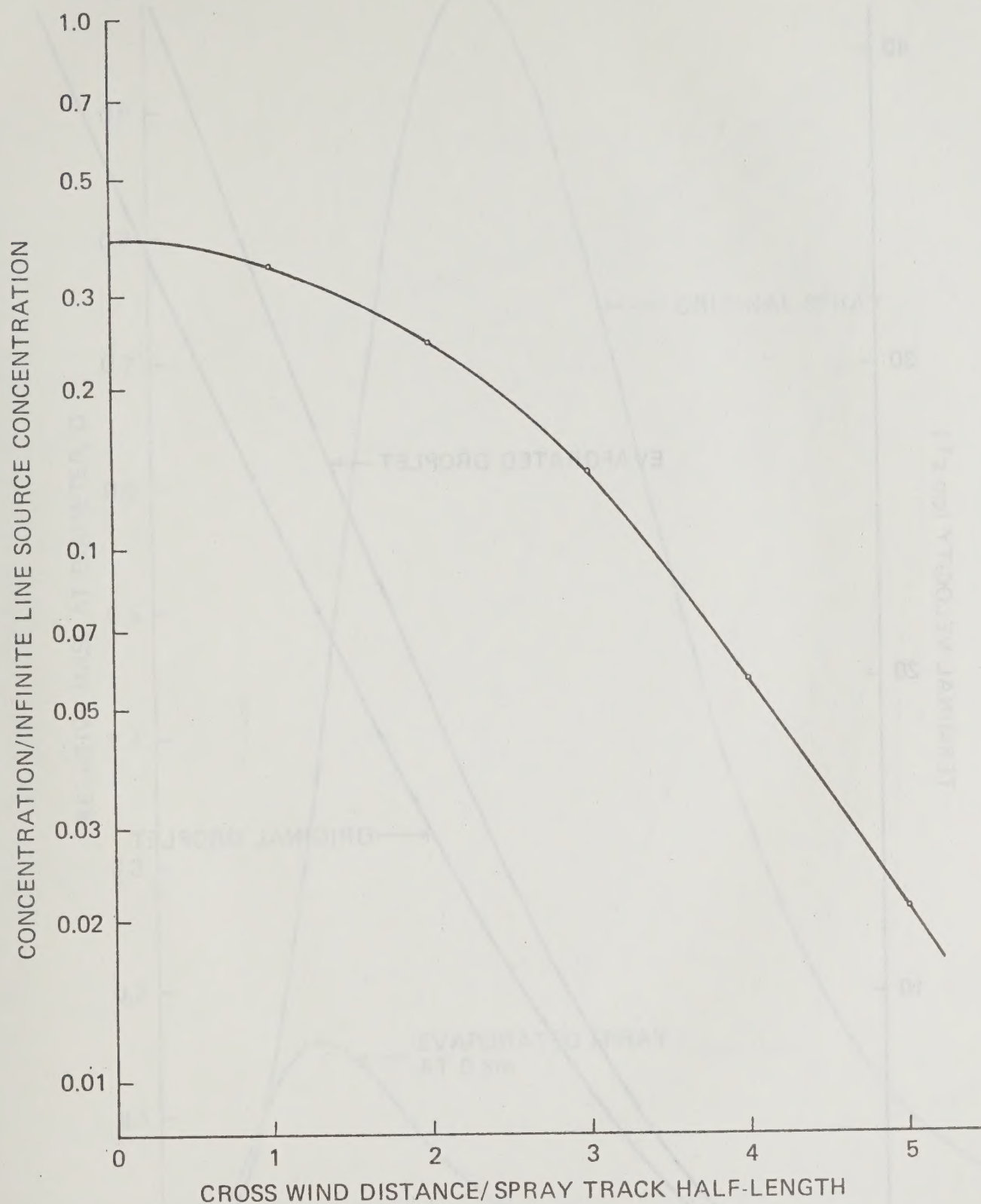


Figure 2: Normalized Concentration at 80 km Downwind in Neutral Conditions as a Function of Normalized Crosswind Distance

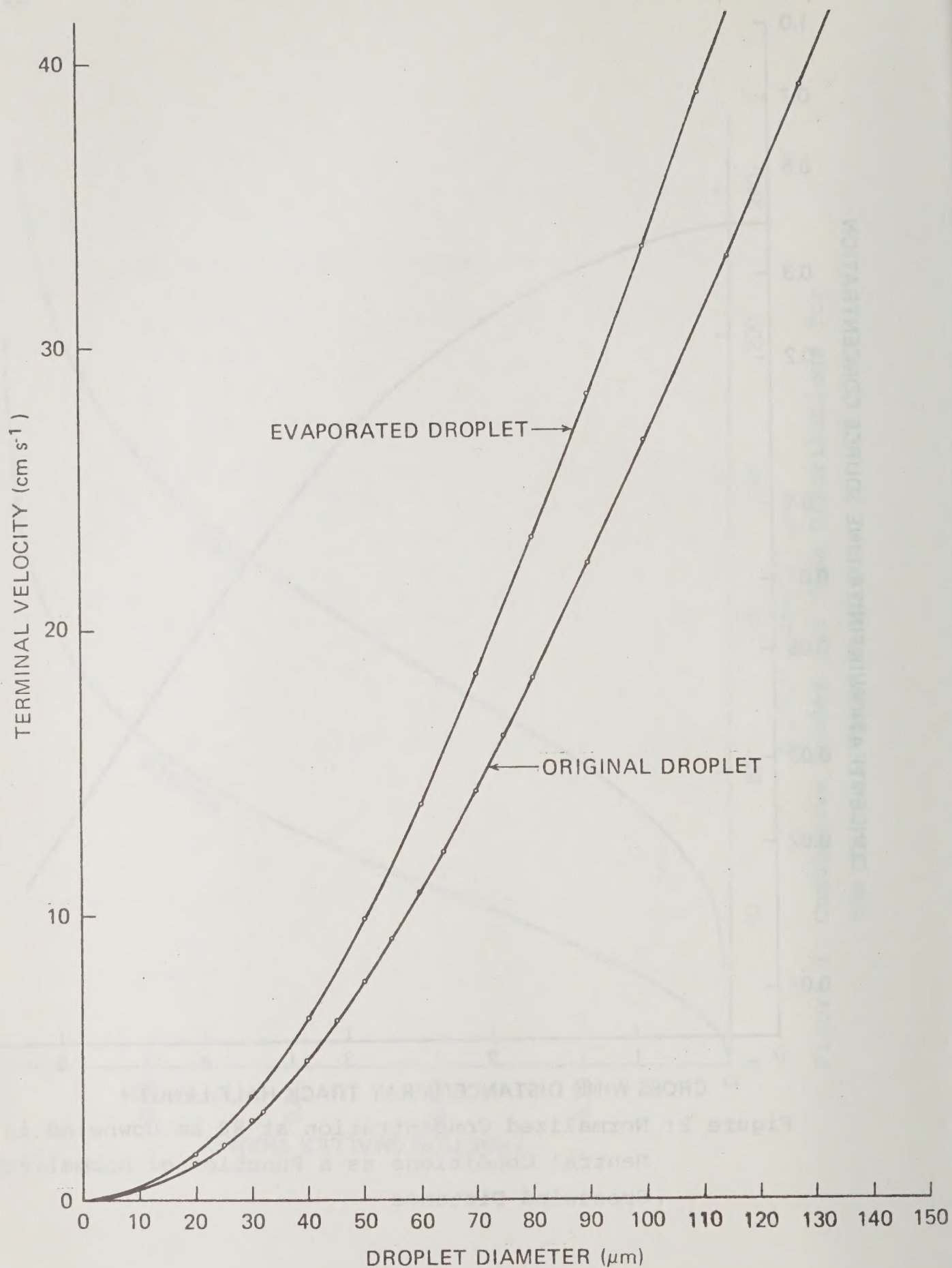


Figure 3: Droplet Terminal Velocity as a Function of Diameter for Original Spray and Evaporated Droplets

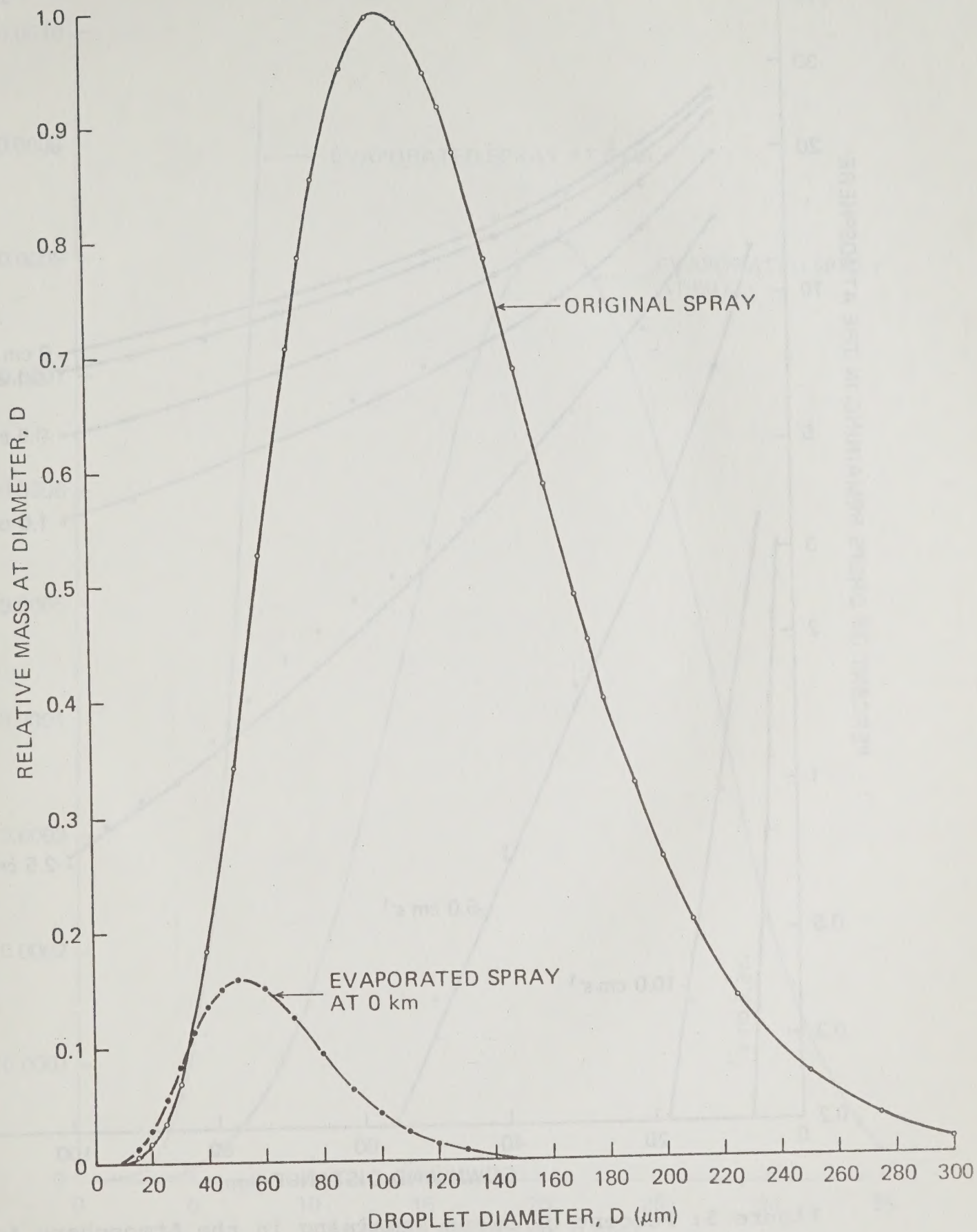


Figure 4: Mass Distribution of Spray Droplets Before and After Evaporation

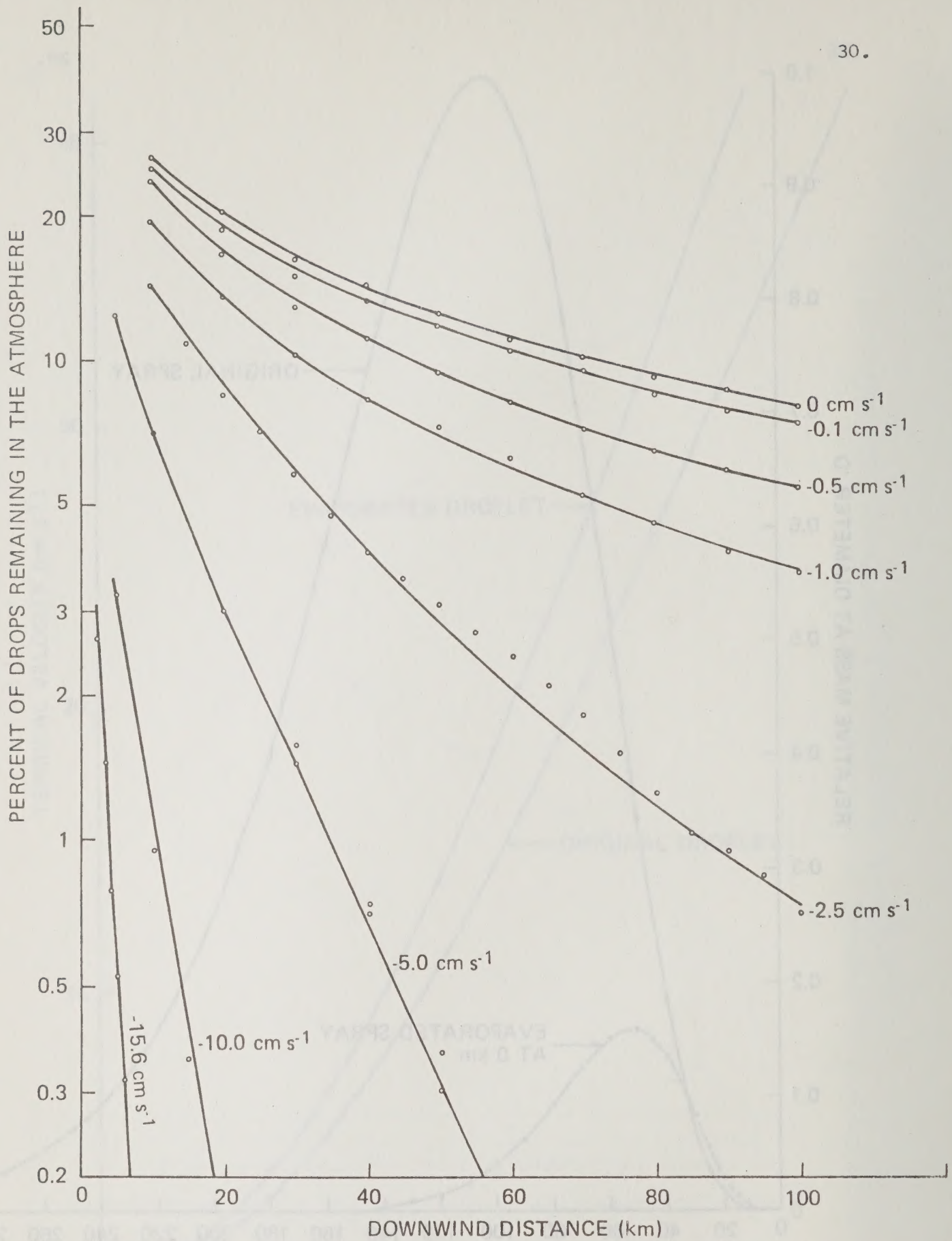


Figure 5: Percent of Drops Remaining in the Atmosphere for various Drop Terminal Velocities as a function of Downwind Distance

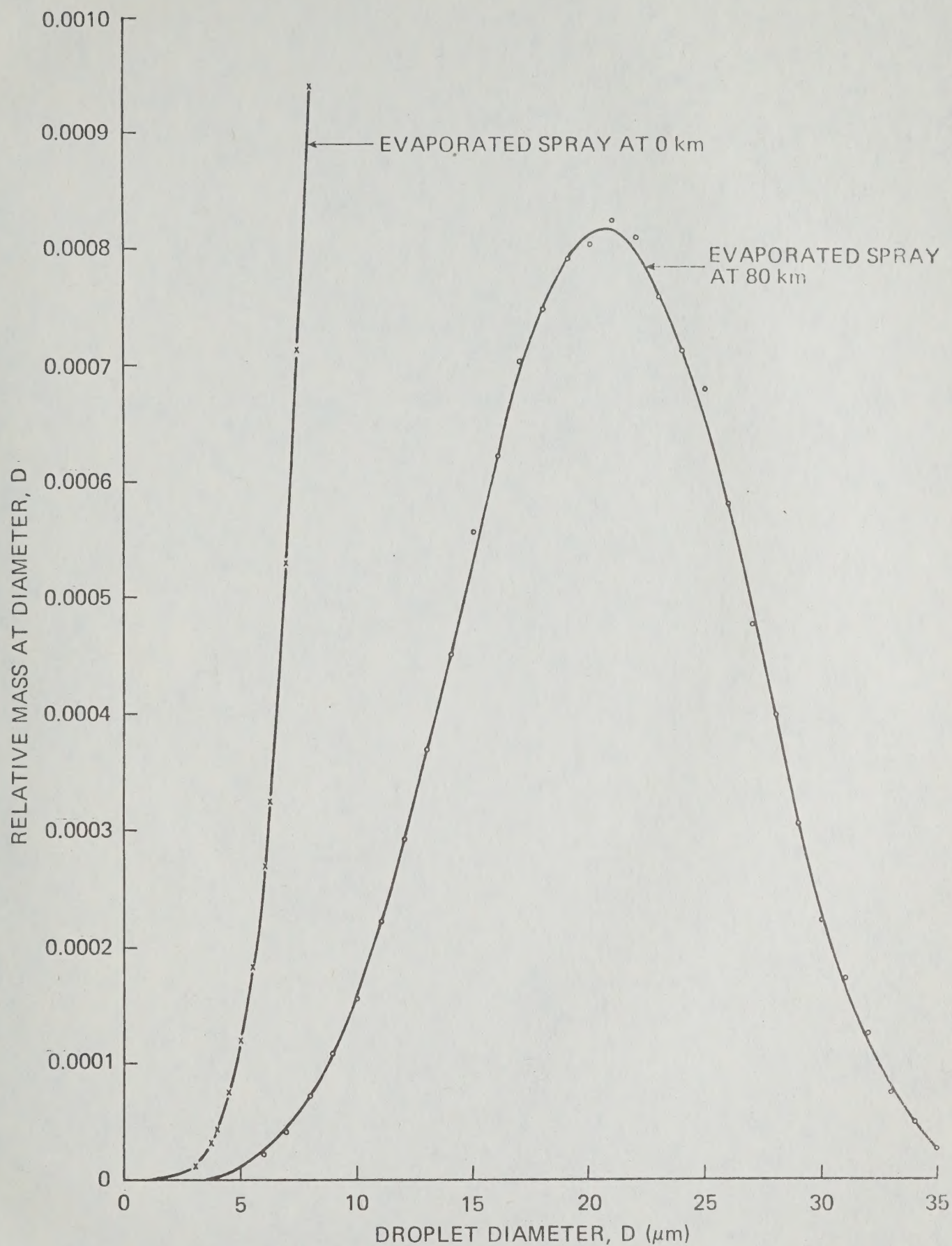


Figure 6: Mass Distributions of Evaporated Spray Droplets at 0 and 80 km Downwind

